



IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re Application of  
Hiroya OKUMURA et al.

Serial No. 09/950,081

Group Art Unit 1745

Filed September 12, 2001

Examiner Mr./Mrs. Mark RUTHKOSKY

For: SEPARATOR FOR SOLID POLYMER TYPE FUEL CELL AND  
PROCESS FOR PRODUCING THE SAME

DECLARATION UNDER RULE 132

Honorable Commissioner of Patent and Trademarks,  
Washington, D.C.

Sir,

I, Koji TAKABATAKE, declare:

That I am a citizen of Japan, residing at 3-6-19-  
311 Nyoidani, Minoo-shi, Osaka, Japan;

That I was born on December 2, 1949 in Okayama and  
graduated from the Department of Synthetic Chemistry,  
Faculty of Engineering Science, Osaka University,  
Toyonaka-shi, Osaka, Japan, 560 in March 1972;

That I have been employed by Nippon Shokubai  
Kagaku Kogyo Co., Ltd. (current NIPPON SHOKUBAI Co.,  
Ltd.), JAPAN since 1972, and engaged in the research in  
thermosetting resins and sheet, and bulk molding  
compounds;

That I have been on loan to Japan Composite Co.,  
LTD. which is a joint venture company of Mitsui Takeda  
Chemicals, Inc. and NIPPON SHOKUBAI Co., Ltd. since  
April 2003, and engaged in the research in separators  
for fuel cell, which was taken over from Mitsui Takeda

Chemicals, Inc., as Senior Chief Researcher since April 2004; and

That the following experiments were conducted under my direct supervision;

#### **EXPERIMENT**

I investigated effectiveness of pressurized kneading on compounds and molded articles thereof.

##### **Sample A**

In accordance with Sample 23648 employing Resin G in Examples of US 6,251,308 (Butler), the sample plate was produced. Incidentally, since Resin G, Modifier B and Graphite A of Sample 23648 were hard to obtain, we used alternate materials instead of the unacquirable components.

With respect to Resin G, Butler refers to that "Resin G is 9100 from Rechhold Chemicals, Inc., ... characterized as a bisphenol-epoxy vinyl ester ... diluted to 54-58 wt% solids with styrene", and the catalog from Rechhold Chemicals, Inc. describes the resin 9100 having 56 wt% solid and 500 cps of viscosity. Therefore, a bisphenol-epoxy vinyl ester resin having similar solid concentration and viscosity was produced.

Incidentally, I enclose the catalog from Rechhold Chemicals, Inc. regarding the resin 9100.

As for an alternate material for Modifier B (polyisocyanate having a NCO content of 17.7 to 20.9), diphenylmethane-4-4'-diisocyanate was employed and the amount added to the resin was adjusted to have the same NCO content (19.3: the medium value of 17.7 to 20.9) with Modifier B.

Regarding an alternate material for Graphite A (having less than 10% greater than 150  $\mu\text{m}$  and less than 10% smaller than 44  $\mu\text{m}$  in diameter), Graphite "SPG 100" having mean particle size of 125  $\mu\text{m}$  was employed, because the mean particle size of Graphite "SPG 100" was between 44 and 150  $\mu\text{m}$ .

That is, to a four-necked flask equipped with a stirrer, a condenser, a nitrogen gas inlet, and a thermometer were charged 956 g of a bisphenol-A-type epoxy resin (manufactured by Toto Kasei Co. Ltd., epototo YD-7011R, epoxy equivalent 478 g/eq), 172 g of methacrylic acid, 0.2 g of triphenylphosphine, 0.1 g of hydroquinone as a thermal polymerization inhibitor, and reacted for 8 hours at 120°C to obtain 1128 g of vinyl ester resin having an acid value of 11.6 mgKOH/g. The vinyl ester resin was diluted with 886 g of styrene monomer to obtain a Resin-G-simulated vinyl ester resin (2014 g). The Resin-G-simulated vinyl ester resin had 56 wt% solid and 520 cps of viscosity. Since the Resin-G-simulated vinyl ester resin was quite similar to Resin G regarding solid concentration and viscosity, we recognized that the Resin-G-simulated vinyl ester resin fell under Resin G.

Further, according to the formulation in Table 1, the molding composition was prepared by adding the resin (Resin-G-simulated vinyl ester resin), monomer initiator (tert-butyl peroxy isopropyl carbonate), inhibitor (2,6-di-tertbutyl-p-cresol), mold release agent (calcium stearate), and rheological modifier (diphenylmethane-4-4'-diisocyanate) to a disperser and

blending for 2 minutes.

To a kneader (manufactured by Toshin Co., Ltd., THM0.5-3 type hybrid mixer, orifice of 10 cm x 9 cm, depth of 15 cm) kneading under an atmospheric pressure, the half amount of conductive filler (Graphite "SPG 100" manufactured by SEC Co. Ltd., mean particle size of 125  $\mu$ m) was added, and with kneading constantly, the above molding composition and the resultant conductive filler were further added and mixed for 15 minutes to obtain a kneaded compound A. When mixing was complete, the kneaded compound A was put in a suitable barrier bag and allowed to mature for approximately one day before molding.

The molding parameters for the molding compositions were as follows: Molding temperature for plaques was 295.degree. F. (146°C) with a molding time of 3 minutes under 100 kgf/cm<sup>2</sup> (approximately 9.8 MPa) to obtain a molded plate A (300 mm x 300 mm x 2 mm).

#### Sample B

A kneaded compound B was obtained in the similar manner to the kneaded compound A except that 110.12 g of Resin-G-simulated vinyl ester resin without the modifier (diphenylmethane-4-4'-diisocyanate) was employed instead of 100 g of the same resin and 10.12g of the modifier, 0.1 g of INT-PS125 (manufactured by Axel Plastics Research Laboratories, Inc.) was used as a release agent instead of 6.0 g of calcium stearate, and kneaded under the pressure of 4 kgf/cm<sup>2</sup> ( $3.92 \times 10^5$

Pa). Incidentally, the ratio of the graphite relative to the resin without the modifier in Sample B is the same with the ratio of the graphite relative to the total amount of the resin and the modifier in Sample A.

For the kneaded compounds and the molded plates obtained in Samples A and B, the following properties were evaluated.

(Kneadability)

The kneadability was visually evaluated by change of compound state at immediately after kneading, and one day after kneading.

(Electric property)

The molded plate was immediately subjected to clip between two billets each 10 mm thick at a room temperature, and allowed to stand for 30 minutes for cooling. The cooled plate was cut for obtaining a test specimen.

The test specimen was subjected to determine the electric property (volume resistivity) according to Japanese Industrial Standards K 7194.

The results are shown in Table 1.

Table 1

	Samples		
	A		B
Pressurized kneading Formulation	Not conducted	conducted	conducted
Resin-G-simulated vinyl ester resin [g]	100	110.12	110.12
diphenylmethane-4-4'-diisocyanate [g]	10.12	0	0
Artificial graphite powder [g]	378.6	378.6	378.6
2,6-di-tertbutyl-p-cresol [g]	0.5	0.5	0.5
tert-butyl peroxy isopropyl carbonate [g]	2.02	2.02	2.02
Mold-releasing agent	Calcium stearate	6.06	0
[g]	INT-PS125	0	0.1
Properties			
Kneadability	Immediately after kneading	Coarse particle	Clay-like
	One day after kneading	Thickened	Clay-like
Electric property (volume resistivity) [mΩcm]		14.4	4.5

### EVALUATION

As apparent from Table 1, the kneaded compound A was coarse particle state immediately after kneading, and thickened one day after kneading. However, the kneaded compound B was clay like state immediately after kneading as well as one day after kneading. Therefore, it is apparent that the pressurized kneading improves the kneading efficiency of the mixture and results in good productivity.

Further, although the ratio of graphite powder relative to resin (or resin and modifier) was the same between Samples A and B, the volume resistivity of Sample A was over three times higher than that of Sample B. Therefore, the pressurized kneading also improves electric conductivity.

I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

Signed this 16<sup>th</sup> day of September, 2005

Koji Takabatake

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